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INTRODUCTION

1.1 PURPOSE

The purpose of this document is to provide a concise overview of the state-of-the-science related to future needs for the development of emission inventories to support planning to reduce high ambient concentrations of PM-2.5, particulate matter with mass median aerodynamic diameter less than 2.5 micrometers (μm). The primary objective is to address issues that are important in the development of emission inventories for PM-2.5 and its precursors. This paper addresses emission factor and activity data development, spatial and temporal variability of PM-2.5, and priorities for methodology and emission factor development on National, regional, and nonattainment area scales. This document is not intended to replace the emissions inventory guidance for PM-2.5 that has been developed by the U.S. EPA.

On May 14, 1999, the U.S. Circuit Court of Appeals for the District of Columbia issued a ruling that remanded the ozone and particulate matter standard, and also called into question what actions, if any, may be taken to implement the new standards. The development of Statewide emission inventories for ozone and particulate matter and their precursors is necessary to address regional issues, irrespective of the final determination on the actual National Ambient Air Quality Standards. Since these are criteria pollutants and are key components of regional haze, development of emission inventories is still deemed to be appropriate including the statewide periodic emission inventories for calendar year 1999. Thus, it is appropriate for the Regions and State/local agencies to:

- discuss plans for developing emission inventories for 1999;
- identify appropriate future actions, programs and associated milestones regarding emission inventories; and
- identify specific issues which need resolution prior to making progress on emission inventories.

1.2 BACKGROUND

1.2.1 *Origins of PM-2.5*

There are three primary origins of PM-2.5: primary solid particulate matter that is emitted directly in the solid phase, primary condensable particulate that can be emitted at high temperature in the gas phase but which condenses into the solid phase upon dilution and cooling in the plume, and secondary particulate that is formed through atmospheric reactions of gaseous SO₂ and NO_x precursor emissions. The secondary components involve complex chemical and physical interactions among the precursor emissions in the atmosphere. Control of the secondary components will require appropriate controls on the sources of the various precursors. The atmospheric reactions and transport processes that result in secondary particulate formation extend over large distances from the sources of the emissions. Both directly emitted and secondary PM-2.5 can be transported over large distances.

Primary PM-2.5 particulate results largely from combustion of fossil fuels or biomass, although selected industrial processes can also be significant in some areas. The sources of PM-2.5 include, but are not limited to, gasoline and diesel exhaust, wood stoves and fireplaces, land clearing, wildland prescribed burning, and wild fires. Sources of primary particulate including fugitive emissions from paved and unpaved roads, dust from ore processing and refining, and to a lesser extent, crustal material from construction activities, agricultural tilling, wind erosion and other crustal source are less important based on their relatively small contribution to ambient PM-2.5 concentrations. The condensable components are largely made up of semi-volatile organic compounds that condense at ambient temperature to form aerosol. The condensable component represents a significant fraction of the PM-2.5 emitted from some sources. Testing for condensables, by means of Method 202, is necessary to characterize PM-2.5 emissions from many sources. EPA is currently exploring other options for source testing methods to characterize the condensable fraction more completely and with greater accuracy.

Secondary PM-2.5 forms through heterogeneous (gas to particle) chemical reactions that convert some common gaseous pollutants into very small particles. The observed secondary PM-2.5 is dominated by sulfur and nitrogen species in most locations, however, there can also be significant contributions from secondary organic aerosol in some locations.

Sulfate aerosol chemistry can involve either gas-phase or liquid-phase oxidation processes. Gas-phase conversion involves the oxidation of sulfur dioxide (SO₂) by hydroxyl radicals (OH), which are formed through the photodecomposition of ozone. The reaction results in sulfuric acid aerosol as an airborne particle with a typical diameter of less than 0.3 μm. The sulfuric acid particles are then neutralized by ammonia and molecular oxygen to form ammonium bisulfate and ammonium sulfate. Liquid-phase reactions involve SO₂ and

oxidants (H_2O_2 , O_3 and O) along with catalysts, such as manganese and iron, that are dissolved in cloud or fog droplets to form sulfuric acid aerosols with diameters in the 0.5 to 1.0 μm size range.

Nitrate aerosols can be produced by several pathways. The most important pathway is the gas-phase reaction of NO_2 with hydroxyl radicals to produce nitric acid, which is then neutralized by gaseous ammonia. When the levels of ammonia and nitric acid are sufficiently high, ammonium nitrate can be formed within the PM-2.5 size range.

Secondary organic aerosols (SOA) are formed by more complex mechanisms involving organic gas-phase particle precursors in the presence of ozone and hydroxyl radicals. The precursors of SOA arise from both anthropogenic and natural sources. Studies have shown that a significant fraction of the α - and β -pinenes, which react with ozone and hydroxyl radicals in the atmosphere, can lead to organic particle formation which then becomes a component of the organic carbon fraction. For example, in poorly ventilated urban areas such as Los Angeles, which has extensive brush covered hills, natural emissions of gaseous hydrocarbons may contribute up to 50% of the secondary organic particles. (NRC, 1993)

Currently, the details of SOA formation are not well known and the implications for needs related to the development of emission factors and other emissions estimation tools to characterize the precursor emissions are uncertain. Large carbon number organic compounds that have an affinity to stick together may contribute significantly to these processes. These processes may also be catalyzed by metals or other trace components. Future development efforts may need to be directed to expand VOC speciation profiles to include compounds that have not been of interest in ozone chemistry and to improve the methods for characterizing sources of trace metals.

These conversion mechanisms from gas to particle forms are thought to be one of the primary atmospheric removal mechanisms for gaseous pollutants. Once in the particle phase, those pollutants are efficiently removed, under the right conditions, by acting as cloud nuclei or through wet and dry deposition to the surface.

1.2.2 Relationship of PM-2.5 to PM-10

PM-10 primary emission inventories are typically dominated by fugitive dust sources. These sources were estimated to contribute approximately 89% of the national total PM-10 emissions in the 1996 National Emissions Trends (NET) inventory. Within the fugitive dust category, the main sources were unpaved roads (33%), wind erosion of natural soils (20%), agricultural crops (17%), construction (14%), paved roads (9%), and all other sources of fugitive PM-10 (2%). (EPA, 1997) However, about 80% of these fugitive dust emissions are larger than 2.5 μm . In all cases, these sources contribute primary particle emissions. In general, the contribution of combustion, condensable and secondary particle origins to PM-10

are an important but usually smaller fraction of the mass, although areas influenced by emissions from woodstoves or other forms of biomass burning and some urban areas can be exceptions.

The available ambient measurements of PM-2.5, however, suggest that anthropogenic combustion sources, fires, and other emitters of condensable and secondary origins contribute a large percentage of the overall PM-2.5 mass in most areas. Data will be needed to evaluate the extent to which existing control programs for PM-10 will benefit PM-2.5. While the effect of existing PM-10 controls on reducing PM-2.5 should be evaluated, planning for PM-2.5 programs may extend to different sources and source categories than those previously targeted for PM-10 programs. In some cases, PM-2.5 planning programs may also benefit from improving the rule effectiveness and compliance with existing pollution control rules for sources of particulates, ozone precursors, and acid rain precursors.

1.2.3 Relationship of PM-2.5 to Visibility Impairment

Visibility impairment is caused primarily by the scattering and absorption of light by fine particles in the atmosphere. Fine particles less than 2.5 μm in diameter scatter and absorb light more efficiently than larger particles. Regional haze is visibility impairment that is produced by a multitude of sources and activities emitting fine particles and their precursors which are located across a broad geographic area. The fine particulate matter (e.g., sulfates, nitrates, organic and elemental carbon, and soil dust) that impairs visibility is among the same particles related to serious health effects and mortality in humans, as well as to environmental effects such as acid deposition and eutrophication.

Data from the Interagency Monitoring of Protected Visual Environments (IMPROVE) network show that visibility impairment in the eastern U.S. is dominated by sulfate, up to 90% for the worst visibility days in some class I areas. In the west, sulfate accounts for about a third of the visibility impairment in many areas, but other components of PM-2.5 such as organic carbon and nitrates contribute more significantly as a percentage of overall visibility impairment levels than in the east. There appears to be a larger contribution from primary emissions from wood smoke and other combustion sources in the west.

The Clean Air Act sets a National goal of remedying existing impairment and preventing future impairment in the 156 Class I National Parks and Wilderness Areas across the country. State implementation plans are to ensure “reasonable progress” toward that goal. Because common pollutants, sources, and transport processes contribute to both health and visibility problems, EPA encourages States to coordinate their planning and assessment activities for PM-2.5 and regional haze programs. The development of statewide PM-2.5 emission inventories will be needed as a foundation for regional planning activities for both PM-2.5 and regional haze.

1.2.4 Organization of the Report

The remaining sections of this report provide a preliminary assessment of some of the important issues that need to be addressed and understood before starting activities to develop an inventory for PM-2.5. The next section discusses the primary sources of PM-2.5 based on available air quality data. This assessment is based primarily on data collected to support analyses of visibility in National Parks and Recreation Areas, and details on specific urban areas are not well represented. A review of the approaches used to develop emission factors and emissions inventories is provided along with a summary of some specific inventory development tools that are available for PM-2.5 programs. The following section summarizes some of the important considerations that might affect PM-2.5 inventory development activities and is meant to be a summary to help States get started on future planning activities. Some selected techniques that can be applied to evaluate the quality or validity of inventory data are presented. The report concludes with some general recommendations that are meant to provide some insights to help in planning and prioritizing efforts in the future. Pie charts showing measured ambient concentrations are shown in Appendix A. Appendix B presents a concise summary of data and tools that are available and remaining needs related to the primary components of PM-2.5. This report is not meant to be a comprehensive guide on inventory development or to identify all of the specific issues that might affect planning in any given area.

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